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Optical bistability in artificial composite nanoscale molecules: Towards all optical processing at the nanoscale

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Abstract. Optical response of artificial composite nanoscale molecules comprising a closely spaced noble metal nanoparticle and a semiconductor quantum dot have been studied theoretically. We consider a system composed of an Au particle and CdSe or CdSe/ZnSe quantum dot and predict optical bistability and hysteresis in its response, which suggests various applications, in particular, all-optical processing and optical memory.

Introduction

Arrays of metallic nano-particles (often referred to as plasmonic arrays), are widely recognized as potential building blocks for nanoscale optical circuits [1, 2, 3]. Recently, a number of papers reported fascinating properties of small clusters of closely spaced semiconductor quantum dots (SQD) and metallic nano-particles (MNP) [4, 5, 6, 7]. Fano resonances [4, 6], bistability in the absorption spectrum [6], and meta- "molecular" resonances [7] have been predicted. When such systems are excited optically, the dipole moment of the excitonic transition in the SQD generates additional electric field at the MNP, which is superposed with the external field, while the induced dipole moment of the MNP generates an additional electric field at the SQD, providing a feedback. Thus, the presence of the MNP leads to a self-action of the optical transition dipole moment, which can give rise to a variety of new optical properties. In particular, if the coupling between two particles is strong the self-action can be large enough to result in optical bistability.

We consider a CdSe or CdSe/ZnSe spherical semiconductor quantum dot and a Au nano-sphere, the simplest artificial diatomic nano-molecule (see the schematic view of the system in Fig. 1). We demonstrate that optical bistability and hysteresis can be observed in the system. The two stable states of the systems have different polarizations, providing a possibility to store information in the form of the system polarization. We argue also that in addition to the traditional way of switching by the amplitude of the driving electric field, the state of such artificial diatomic molecule can be switched by the polarization of the field with respect to the molecule axis. The fact that both the SQD and the MNP can sustain high electric fields suggests such possible applications of the artificial molecule as an all-optical switch and optical memory at nano-scale.

Formalism

We assume that the SQD-MNP system is embedded in a dielectric host with the permeability ε_b and driven by the external electric field with the amplitude $\mathbf{E}_0$ and frequency ω. The SQD is treated quantum mechanically (as a two level system) within the framework of the Maxwell-Bloch equations while the MNP is treated classically; the response of the MNP is described by its frequency dependent polarizability within the point dipole approximation. The rotating wave approximation is used throughout the paper, so that all time dependent quantities represent amplitudes of the corresponding signals, the set of equations for which reads:

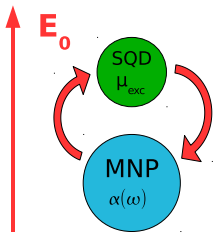

Fig. 1. Schematics of a semiconductor quantum dot - metallic nanoparticle hybrid system embedded into a homogeneous dielectric background with permeability ε_b and subjected to an external field with the amplitude $\mathbf{E}_0$.

$$\dot{Z} = -\gamma\,(Z+1) - \frac{1}{2}\,(\,\Omega\,R^* + \Omega^* R\,)\ , \tag{1}$$

$$\dot{R} = -(i\,\Delta + \Gamma)\,R + \Omega\,Z\ , \tag{2}$$

where $Z = \rho_{11} - \rho_{00}$ is the population difference, R is the amplitude of the off-diagonal density matrix element ρ_{10}, γ and Γ are relaxation rates, Δ is the detuning of the SQD resonance and $\Omega = \boldsymbol{\mu}\mathbf{E}/\hbar$ is the Rabi frequency ($\boldsymbol{\mu}$ is the optical transition dipole moment of the SQD). The total electric field $\mathbf{E}$ at the SQD is the superposition of the external field $\mathbf{E}_0$ and the scattered field produced by the MNP:

$$\mathbf{E} = \frac{1}{\varepsilon_{\rm s}'}\left(\mathbf{E}_0 + \frac{\bar{\mathbf{S}}\,\mathbf{P}_{\rm MNP}}{\varepsilon_b\,d^3}\right)\ . \tag{3}$$

Here, $\varepsilon_{\rm s}' = 3\varepsilon_{\rm b}/(\varepsilon_{\rm s}+2\varepsilon_{\rm b})$, $\varepsilon_{\rm s}$ is the SQD dielectric constant, $\bar{\mathbf{S}} = {\rm diag}(-1,-1,2)$ is the angular part of the dipole field Green tensor, d is the SQD-MNP center-to-center distance and $\mathbf{P}_{\rm MNP}$ is the induced dipole moment of the MNP:

$$\mathbf{P}_{\rm MNP} = \alpha(\omega)\,a^3\left(\mathbf{E}_0 + \frac{\bar{\mathbf{S}}\,\mathbf{P}_{\rm SQD}}{\varepsilon_b\,d^3}\right)\ , \tag{4}$$

where $\alpha(\omega)a^3$ is the classical frequency dependent polarizability of the MNP, a being its radius and $\alpha(\omega) = [\varepsilon_M(\omega) - \varepsilon_b]/[\varepsilon_M(\omega) + 2\varepsilon_b]$ with $\varepsilon_M(\omega)$ being the dielectric function of the metal while $\mathbf{P}_{\rm SQD} = (-i/2)R\,\boldsymbol{\mu}$ is the SQD dipole moment amplitude. The total electric field in the SQD is therefore given by

$$\mathbf{E} = \frac{1}{\varepsilon_{\rm s}'}\left[\mathbf{1} + \frac{\alpha(\omega)\,a^3}{\varepsilon_b\,d^3}\,\bar{\mathbf{S}}\right]\mathbf{E}_0 + \frac{\alpha(\omega)\,a^3}{\varepsilon_{\rm s}'\varepsilon_b\,d^6}\,\bar{\mathbf{S}}^2\,\mathbf{P}_{\rm SQD}\ . \tag{5}$$

The Rabi frequency $\Omega = \Omega_0 - i\,G\,R$ entering Eq. (1) and (2) contains the part corresponding to the renormalized external field, Ω_0 [the first term in Eq. (5)], and the self action [the second term in Eq. (5)]. The steady-state equation for the population difference Z reads:

$$\frac{|\Omega_0|^2}{\gamma\,\Gamma} = -\frac{Z+1}{Z}\,\frac{|\Gamma + i\,(\Delta + G\,Z)|^2}{\Gamma^2} \qquad (6)$$

For some set of parameters and values of the driving fields this equation can have three real solutions, only two of which turn up to be stable.

Results

Hereafter we consider spherical CdSe/ZnSe SQD and Au MNP and use the following set of parameters: $\hbar\omega = 2.36$ eV (which corresponds to optical transition in 3.3 nm SQD), $\varepsilon_s = 6.2$, $a = 10$ nm, $d = 17$ nm, $\varepsilon_b = 1$, $1/\gamma = 0.8$ ns, $1/\Gamma = 0.3$ ns, $\Delta = 0$. We use tabulated dielectric function of gold [8] to calculate the polarizability of the MNP.

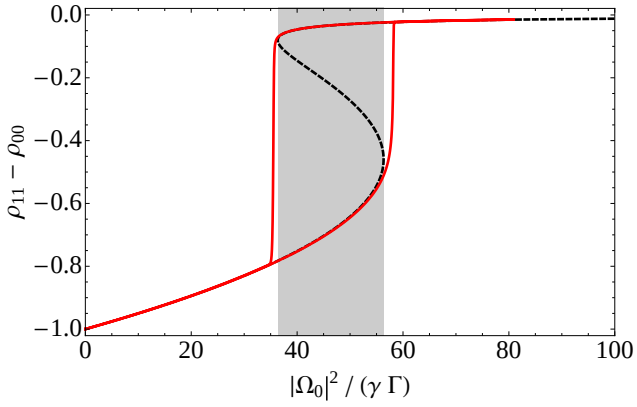

Fig. 2. Dependence of the population difference of the SQD on the renormalized external field Ω_0 manifesting optical bistability (shaded region). Dashed line – the three-valued solution of Eq. (6). Solid line – solution of Eq. (6) with time dependent external field sweeping back and forth across the bistability region.

Dashed line in Fig. 2 shows the solution to Eq. (6), the dependence of the steady state population difference on the renormalized external field Ω_0 (the field is parallel to the system axis). Shaded region shows the range of the external field for which the system can have three different states. Only the upper and lower branches of this solution are stable, resulting in optical bistability. To observe the two branches, we sweep the external field back and forth across the bistability region. Solid line in Fig. 2 shows the result of the corresponding calculation. The figure demonstrates that when the field is increasing (from 0), the population difference follows the lower stable branch until the critical field amplitude is reached. Further, the system switches abruptly to its other stable state (upper branch). When we sweep the field back the upper-to-lower branch switch occurs at a different critical field, forming the typical hysteresis loop.

The bistability of the population of the SQD results in the bistability of SQD and MNP polarizations. In Fig. 3, we present the hysteresis loop of the polarization of the SQD calculated for the same parameters as in Fig. 2. The two

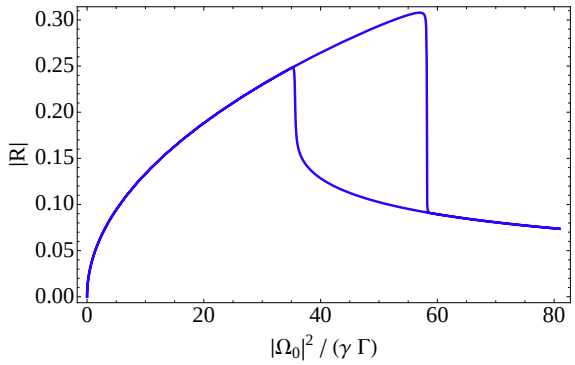

Fig. 3. Dependence of the dipole moment amplitude $|R|$ of the SQD on the renormalized external field Ω_0, manifesting optical bistability in the polarization of the SQD.

stable states of the SQD population correspond to two different polarization, providing the possibility for storage of information by the system polarization.

Finally, we point out that the specific feature of the SQD-MNP system is the existence of the symmetry axis; this allows us to drive the system not only by the amplitude of the external field, but also by the orientation of its polarization with respect to the axis. These are new properties which can be useful for applications.

Summary

We investigated theoretically the optical response of a hybrid "artificial" molecule composed of a semiconductor quantum dot, modeled as a two-level system and a metal nanoparticle, considered classically, which are coupled by the dipole-dipole interaction. The interaction results in a self-action of the SQD via MNP, leading to the dependence of the SQD optical transition frequency on the population, which provide a feedback mechanism resulting in several fascinating effects. Thus, in the strong coupling regime, we found that the system can manifest bistability and optical hysteresis, as well as switching of the polarization of both SQD and MNP by the incoming field. Such switching can be achieved not only by the traditional amplitude control but also by the polarization of the incoming field with respect to the system axis; the latter being very promising for optical memory applications at the nanoscale.

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